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# IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

Werner POMPETZKI et al

: GROUP ART UNIT: 1621

SERIAL NO: 09/618,044

FILED: July 17, 2000

: GROUP ART UNIT: E. Price

FOR: PROCESS FOR THE

HYDROGENATION OF ACETONE

# **APPEAL BRIEF**

ASSISTANT COMMISSIONER FOR PATENTS WASHINGTON, D.C. 20231

SIR:

The following is an appeal to the Board of Appeals concerning the final decision by the Examiner to reject Claims 1-3 and 5-17 of the above-identified application.

# **REAL PARTY OF INTEREST**

Degussa-Huls is the real party of interest of the present application.

## RELATED APPEALS AND INTERFERENCES

# **STATUS OF CLAIMS**

Claims 3 is canceled by way of amendment filed herewith. Claim 4 has been canceled. Claims 1, 2 and 5-17 remain active in the case.

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# **STATUS OF AMENDMENTS**

No response to the final Office Action dated January 22, 2002 has been filed.

# **SUMMARY OF THE INVENTION**

The present invention is directed to a process for the hydrogenation of acetone by conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by weight in at least two hydrogenation process stages, thereby preparing isopropanol product.

Support for the invention as summarized can be found on page 2, lines 24-28 and page 5, lines 10-11 of the specification.

# **ISSUES**

Whether Claims 1-3 and 5-17 stand properly rejected based on 35 USC 103(a) as obvious over Fukuhara et al, U. S. Patent 5,081,321 in view of Hiles et al, U. S. Patent 4,626,604.

# **GROUPING OF CLAIMS**

Claims 2 and 5-11 and 13-15 do not stand or fall together with Claim 1.

# **ARGUMENT**

The process of the present invention is directed to the hydrogenation of acetone by conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by wt in at least two hydrogenation process stages thereby preparing the

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desired isopropanol product in high yield and selectivity with substantially reduced by-product production. The two essential features of the present process which enable the superior results of the hydrogenation of acetone are that (1) the reaction is conducted in at least two successive hydrogenation stages and that (2) the hydrogenation of acetone is conducted under the condition of the acetone having a water content of  $\leq 1$ % by weight. In this regard Appellants point out that they have presented clear evidence in the record of the case of the importance or criticality of these two process aspects of the present invention. That is, the Example of the present application on pages 8 and 9 shows the hydrogenation of acetone in two process stages using a nickel catalyst (10 % by weight Ni on a neutral ( $\alpha$ -Al<sub>2</sub>O<sub>3</sub> support). The isopropanol product is obtained in a high yield of 99.45 % while the amount of by-products is less than 100 ppm at the outlet of the second hydrogenation stage. Thus, there is a clear demonstration in the specification of the essential features of the present process which result in the production of isopropanol product in high yield and purity.

Appellants have also submitted evidence into the record in the form of the Declaration (37 CFR 1.132) executed July 4, 2001 which shows the importance of the no more than 1 % by wt water content limitation in acetone starting material of the present claims and its impact on isopropanol product purity. The data in the table of the declaration, a copy of which is attached to the brief, clearly show that as the water content of acetone starting material increases, the amounts of by-product impurities in the product isopropanol continue to increase. Such a trend is clearly not shown or suggested by the cited prior art.

As to the <u>Fukuhara et al</u> patent, the same shows the hydrogenation of acetone to isopropanol product in a single reactor. The patent is absolutely silent as to the essential limitations of the present claims of multiple hydrogenation stages, the percentage of water

contained in the acetone reactant being limited to ≥ 1 % and the amount of by-products if any. In fact, the Examiner has remarked on the record that in deed the Fukuhara et al patent does not teach or suggest these essential features of the present claims. Appellants therefore question, in light of these admissions, why the present claims have been rejected over Fukuhara et al, because it is precisely these three features that define the present invention?

Appellants have demonstrated on the record that the improved results provided by the present

of water in at least two hydrogenation stages.

Considering the disclosure of Fukuhara et al in more detail, the reference discloses an entirely different process of hydrogenating acetone as described at the bottom of column 2 through the top of column 3. The essential features of the hydrogenation process of the reference are that it is relegated to a fixed bed reaction system having a granular or particulate catalyst therein. In this reactor configuration, it is essential to conduct the direction of flow of reactant liquid and hydrogen gas over the catalyst in a specific direction. That is, it is critical to provide a co-current liquid/gas down-flow relative to the fixed bed catalyst and to maintain the catalyst bed in a trickle bed state. These features which are critical to the process of Fukuhara et al are irrelevant as far as the process of the present invention is concerned. As is clear from the description above concerning the present invention, the important features of the hydrogenation process are that the acetone reactant contain no more than 1% by weight water and that the hydrogenation process be conducted in at least two hydrogenation stages. Accordingly, the Fukuhara et al reference appears to be of secondary interest.

invention are only achieved by conducting the hydrogenation of acetone that contains ≥ 1 %

With respect to the requirement of the present claims of conducting the two stage hydrogenation of acetone having a water content of no more than 1% by weight, the Examiner states at page 5, lines 2-6 that there is no reason for him to believe anything other than that patentees used an acetone starting material which meets the requirement of the present invention of an acetone starting material containing  $\geq 1^{-}\%$  of water. This statement is made in light of the clear fact that patentees have stated nothing in their disclosure as to the water content of the starting acetone material and the fact that, as stated on page 5, lines 6-9 of the text, the relevant prior art, in fact, has added water to acetone feed streams to increase selectivity or to reduce by-product formation. Why then would it be expected that the acetone starting material of  $\underline{Fukuhara\ et\ al}$  would necessarily contain no more than 1 % water? In this connection the Examiner states that a commercially available acetone contains less than 1 % water. Certainly, acetone product is and has been commercially available which contains less than 1 % water. But this availability in no way suggests that such a low water content acetone would be the obvious and necessary choice by one of skill in the art for a two stage hydrogenation process (not taught by Fukuhara et al) in the large scale conversion of acetone to isopropanol.

As to the disclosure of <u>Hiles et al</u>, it is not clear to Appellants how the <u>Hiles et al</u> reference can be properly combined with the disclosure of <u>Fukuhara et al</u>, because the two patents are directed to two quite different aspects of aldehyde (ketone) hydrogenation. The aspect of improvement of the catalytic hydrogenation of aldehydes which <u>Hiles et al</u> have achieved is to conduct catalytic hydrogenation in at least three hydrogenation zones of which a first is a first catalytic stage, the second is a hydrogenation and at least one intermediate stage including a penultimate stage, followed by a final hydrogenation step. Each stage

contains a charge of hydrogenation catalyst and the stages are connected in series so that the material from one stage is fed to the next stage in series. Other factors of the multistage hydrogenation process are disclosed in column 4 of the reference. Note in this regard that the Hiles et al reference specifically comments on the inadequacies of a plural stage hydrogenation where hydrogenation is conducted in only two stages. Apparently in a twostage process the operator of a hydrogenation process would not be able to compensate adequately for a decline in catalyst activity without making certain adjustments to the process (column 5, lines 32-45). Accordingly, if there are in fact inadequacies of a two stage hydrogenation as stated by Hiles et al, and their solution is to provide a hydrogenation process of at least three stages, how could such a disclosure lead the skilled artisan to the present invention, where although more than two hydrogenation stages can be employed in the hydrogenation process, nevertheless a two stage process is adequate with the proviso that the water content of the acetone starting material be no more than 1 % by weight? Clearly there is no teaching or suggestion in Hiles et al of this critical feature of the present process, so that, whereas Hiles et al would lead the skilled artisan to conclude that a two stage hydrogenation of acetone is inadequate, to the contrary, in the present invention, at least two hydrogenation stages or steps are adequate as long as the water content limitation set forth in the present claims is maintained.

Appellants submit that to one of skill in the art desiring to improve the selectivity of a multistage hydrogenation process as disclosed in <u>Hiles et al</u>, would use, in view of the disclosure of <u>Fukuhara et al</u>, a trickle bed reactor under the very specific flow conditions taught in the <u>Fukuhara et al</u> reference. However, as pointed out above, no motivation is provided to adjust the water content of the acetone starting material in event acetone is

selected for the hydrogenation process. Clearly, not only does <u>Fukuhara et al</u> teach a process quite different from that of the present invention, likewise the <u>Hiles et al</u> reference fails to teach or suggest the invention, and the combined patents still impart no motivation whatsoever to the skilled artisan to hydrogenate a starting acetone material containing no more than 1 % by wt water in at least two hydrogenation stages with the expectation of obtaining a highly pure isopropanol product in high yields.

As to the matter, as stated at the bottom of page 5 of the final Office Action, that in view of the availability of low water content acetone as offered for sale by "Sigma," the Examiner is advised to consider that the offering of sale of products from a specialties chemical supply source such as Sigma is a clear indication of the sale of a material in relatively small volume amounts and not in the much larger amounts required by industry for a process such as that of the present invention where comparatively vast amounts of acetone are required for a given process. Accordingly, the combination of the two cited patents is not believed to suggest the present invention.

## Claim 2

Given that neither cited patent of record teaches or suggests the limitations of present Claim 1, neither reference obviously teaches or suggests the limitations of Claim 2 of conducting the hydrogenation of acetone in two stages, wherein the first stage is conducted at a temperature of 60 to 140° C and at a pressure of 20 to 50 bar. Withdrawal of the rejection of the claim is respectfully requested.

# Claims 5 and 6

Claims 5 and 6 stand separately patentable because it is clear that because the cited patents do not teach the no more than 1 % water limitation of Claim 1, the patents could not possibly teach the narrower limitations of Claims 5 and 6. Withdrawal of the rejection of the claim is respectfully requested.

# Claims 7 and 8

With respect to a preferred aspect of the present invention in which the hydrogenation catalyst employs a neutral support, it is noted that whereas Fukuhara et al in column 2, lines 20-32 disclose several hydrogenation catalysts, although alumina supports are mentioned, none of the disclosed supports are neutral supports. In this connection it should be observed that it is known that alumina exists in several different modifications, but only  $\alpha$ -A1<sub>2</sub>O<sub>3</sub> is a neutral support as disclosed in the present specification at page 7, line 14. As disclosed in Example 1 of the Fukuhara et al reference, Raney nickel is used, which is developed in an aqueous sodium hydroxide solution. Thus it cannot be concluded from Fukuhara et al that a neutral support material for a catalyst is contemplated.

# Claim 9

The cited and applied patents do not show or suggest a two stage acetone hydrogenation process in which the hydrogenation is conducted at a temperature of 60 to 140° C and a pressure of 25 to 35 bar. Withdrawal of the rejection is respectfully requested.

# Claims 10 and 11

Neither of the cited patents teaches or suggest an at least two stage acetone hydrogenation process, where, in the second stage of hydrogenation, the temperature of the second stage of hydrogenation is 60 to 140° C and a pressure of 20 to 50. Moreover, the narrower temperature range of 70 to 130° C of Claim 11 for the second stage of hydrogenation is neither taught of suggested. Withdrawal of the rejection is respectfully requested.

# **Claims 13 -15**

Neither of the cited patents teaches anything about the amount of impurities formed during the reduction of acetone to isopropanol product. Certainly neither reference provides a teaching of a two stage hydrogenation of acetone starting material which results in an impurity content in product acetone of 300 ppm or less. Withdrawal of the rejection of the claim is respectfully requested.

In view of the comments above, appellants continue to be of the opinion that the continued rejection of the claims of the application is erroneous and should be REVERSED.

Respectfully submitted,

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# **FINDINGS OF FACT**

- 1. The cited and applied Fukuhara et al patent discloses a process of catalytically hydrogenating acetone by feeding hydrogen gas and acetone liquid into a reactor containing a fixed catalyst bed from the top of the reactor to form a co-current gas/liquid downflow.

  There is no teaching of suggestion of a two stage hydrogenation process on a starting acetone material which contains no more than 1 % by wt water. Indeed the Examiner has acknowledged such on the record.
- 2. The cited and applied Hiles et al patent discloses a continuous multi-stage hydrogenation process of reducing unsaturated organic starting materials including aldehydes, an important step of which is controlling the rate of supply of aldehyde or other unsaturated organic in the penultimate stage of hydrogenation in relation to catalyst volume so that the degree of hydrogenation in that stage is less than 100 %. Such a process does not represent the present process in which acetone starting material containing no more than 1 % water is hydrogenated in an at least two stage process resulting in isopropanol product of high purity.



# **CONCLUSION OF LAW**

1. Claims 1, 2 and 5-17 are not rendered obvious under 35 USC 103(a) by the cited combination of Fukuhara et al, U. S. Patent 5,081,321 in view of Hiles et al, U. S. Patent 4,626,604.

# **APPENDIX**

# **CLAIMS ON APPEAL**

1. A process for the hydrogenation of acetone, which comprises:

conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by weight in at least two hydrogenation process stages, thereby preparing isopropanol product.

- 2. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation in a first stage is conducted at a temperature of 60 to 140° C and a pressure of 20 to 50 bar.
- 5. The process as claimed in Claim 1, wherein the acetone to be hydrogenated has a water content of less than or equal to 0.5 % by weight.
- 6. The process as claimed in Claim 1, wherein the acetone to be hydrogenated has a water content of less than or equal to 0.2 % by weight.
- 7. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation is conducted in the presence of a nickel containing catalyst on a neutral support.
  - 8. The process as claimed in Claim 7, wherein said neutral support is  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>.
- 9. The process as claimed in Claim 2, wherein the liquid-phase hydrogenation is conducted at a temperature of 70 to 130° C, and a pressure of 25 to 35 bar.
- 10. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation in a second stage is conducted at a temperature of 60 to 140° C and a pressure of 20 to 50 bar.
- 11. The process as claimed in Claim 10, wherein the liquid-phase hydrogenation is conducted at a temperature of 70 to 130° C.
- 12. The process as claimed in Claim 1, wherein the hydrogenation is conducted at a molar ratio of hydrogen to acetone ranging from 1.5:1 to 1:1.

- 13. The process as claimed in Claim 1, wherein the total concentration of by-products formed in said liquid-phase hydrogenation reaction is less than 300 ppm.
- 14. The process as claimed in Claim 13, wherein said total amount of by-products is less than 200 ppm.
- 15. The process as claimed in Claim 14, wherein said total amount of by-products is less than 100 ppm.
- 16. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation reaction is conducted in the presence of a hydrogenation catalyst of copper, chromium, ruthenium or nickel on a Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> or ZrO<sub>2</sub> support.
- 17. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation reaction is conducted in the presence of a hydrogenation catalyst of a catalytically active metal on a neutral support selected from the group consisting of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub> or mullite.

# TABLE

Product Composition		A	æ	၁	Q	E	Ā	g	Н	I	r
Isopropanol	%	99.92	26.66	99.91	68'66	99.85	78.66	78.66	99.90	68.66	98.66
Acetone	mdd	497	435	461	200	553	724	708	450	575	776
Diisopropylether	mdd	66	142	175	191	198	200	208	202	200	200
Methanol	mdd	36	42	46	49	50	51	51	50	51	52
Mesityloxide (MOX)	mdd	4	3	3	3	3	3	2	2	2	2
Methylisobutylketone (Mibk)	mdd	•	t	. 1	ı	1		1		•	
4-Methylpentanol-2	mdd	9	11	12	-17	20	31	26	26	29	30
Hexylenglycol	mdd	46	86	125	210	175	193	203	196	198	204
Water	mdd	365	464	929	716	753	126	586	1148	1165	1248
Water formation from the starting material	шаа	ca. 124	ca. 163	ca. 91	ca. 155	ca. 73	ca. 222	ca. 195	ca. 107	ca. 119	ca. 149
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